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Classical Ring-exchange Processes on the Triangular Lattice ⋆

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Abstract

The effects of the ring-exchange Hamiltonian $H_3 = J_3 \sum_{\langle ijk\rangle}(S_i \cdot S_j)(S_i \cdot S_k)$ on the triangular lattice are studied using classical Monte Carlo simulations. Each spin S_i is treated as a classical XY spin taking on Q equally spaced angles (Q-states clock model). For $Q = 6$, a first-order transition into a stripe-ordered phase preempts the macroscopic classical degeneracy. For $Q > 6$, a finite window of critical phase exists, intervening between the low-temperature stripe phase and the high-temperature paramagnetic phase.

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The importance of higher-order ring-exchange processes in low-dimensional magnets and its potential role in stabilizing liquid-like exotic ground states has recently been under intense investigation[1,2]. Generically the Hamiltonian is of the type

$$H = \sum_{n\geq 2} H_n \tag{1}$$

with H_n for $n = 2, 3, 4$ given by

$$\begin{aligned} H_2 &= J_2 \sum_{\langle ij\rangle} S_i \cdot S_j \\ H_3 &= J_3 \sum_{\langle ijk\rangle} (S_i \cdot S_j)(S_i \cdot S_k) \\ H_4 &= J_4 \sum_{\langle ijkl\rangle} (S_i \cdot S_j)(S_k \cdot S_l). \end{aligned} \tag{2}$$

Each S_i is a Heisenberg spin, and $\langle ij\rangle$, $\langle ijk\rangle$ and $\langle ijkl\rangle$ refer to nearest-neighbor pair, triplet, and quartet of sites, respectively.

In particular the possibility to realize a stable spin-liquid ground state due to H_3 in a two-dimensional triangular lattice has been suggested in the variational Monte Carlo study of Motrunich[2]. It may be inferred that the three-site exchange process has the "frustrating" effect which renders the liquid ground state energetically more stable over a $\sqrt{3} \times \sqrt{3}$ magnetically ordered structure.

In this paper, we report the first results of isolating the effects of the three-site exchange process by studying the following model. First, we consider the classical counterpart of the Hamiltonian (1) where S_i is treated as a unimodular vector. Secondly, only $H = H_3$ is considered in this paper while leaving the study of the compound models such as $H = H_2 + H_3$ or $H = H_2 + H_3 + H_4$ for the future. Thirdly, we consider the planar spin $S_i = (\cos\theta_i, \sin\theta_i)$. The angle θ_i is divided up into Q equally spaced segments. The same strategy had been applied for $H = H_2$ as a way to asymptotically approach the behavior of the XY model ($Q = \infty$) and is known as the Q-states clock model[3,4].

Antiferromagnetic Q-states models on a triangular lattice have double transitions of XY- and Ising-types with extremely close critical temperatures[5]. The same subtlety might pervade the $H = H_3$ model too, but here we choose to focus on the broader issue: *What is the nature of the low-temperature phase exhibited by $H = H_3$?* The results for $Q = 2$ and $Q = 6$ are discussed in detail in this paper. Some preliminary specific heat data for larger Q are presented.

Q=2: It turns out that $Q = 2$ model maps onto the antiferromagnetic Ising model on the triangular net, first

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studied by Wannier[8]. In the Ising case spins take on $S_i = \pm 1$, and the Hamiltonian H_3 reduces to

$$(S_i \cdot S_j)(S_i \cdot S_k) \to S_j S_k, \quad H_3 \to 2J_3 \sum_{\langle ij \rangle} S_i S_j, \qquad (3)$$

which is the antiferromagnetic Ising model. This model possesses macroscopic degeneracy[8,9] which is also revealed as the residual entropy S_0. Our Monte Carlo (MC) calculation gives $S_0 \approx 0.323 k_B$, in excellent agreement with the value predicted earlier[8,9]. There is no long-range order down to zero temperature in this model.

With $Q \geq 3$ H_2 and H_3 are no longer equivalent. The lowest-energy configurations for H_3 consist of two-up and one-down (or vice versa) spins for each elementary triangle. Thus, macroscopic degeneracy is a general feature of $H = H_3$ for an arbitrary even integer Q. It is also well known that the magnetic ordering for $H = H_2$ is obtained for angles of 120° between nearest-neighbor spins. Such situations are possible if Q is a multiple of 3. To allow the realization of both, we consider the case $Q = 6$.

Q=6: With $Q = 6$ we observed a *first-order* transition at $T_c/J_3 \approx 1.05$. Hysteresis in the average energy and the order parameter (defined below) in our MC runs vindicate the first-order nature, as shown in Fig. 1.

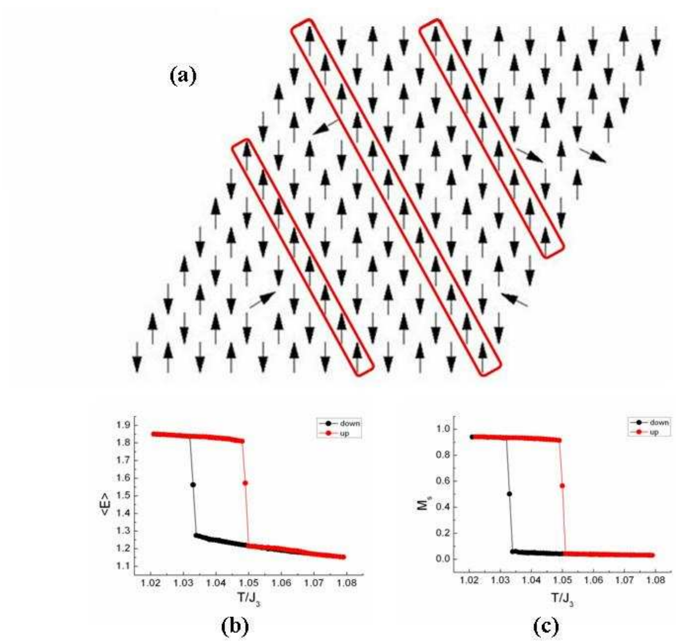

Fig. 1. Low-temperature configuration for the $Q = 6$ model, $H = H_3$. Ferromagnetic ordering within a diagonal stripe and antiferromagnetic ordering between adjacent stripes are apparent in (a). Both the average energy (b) and the average magnetization (c) data are consistent with the first-order phase transition to the low-temperature phase.

The nature of the low-temperature, ordered phase is clearly demonstrated in Fig. 1. We find the spontaneous emergence of stripe-like domains of up and down spins below T_c. The ground state degeneracy is lifted through the order-from-disorder mechanism[6,7]. While a conventional order-from-disorder idea predicts the gradual separation of the free energies of different classical ground state configurations with rising temperatures and no phase transition, our model exhibits a first-order transition which pre-empts the classical macroscopic degeneracy. The difference is due to the discrete nature of our model. The order parameter appropriate for this stripe-like configuration is

$$m = \frac{1}{N} \left| \sum_i (-1)^{i_1} S_i \right|, \qquad (4)$$

where each lattice site is given the coordinate $i = i_1 \hat{e}_1 + i_2 \hat{e}_2$, $\hat{e}_1 = \hat{x}$, $\hat{e}_2 = -\hat{x}/2 + \sqrt{3}\hat{y}/2$, and N is the number of sites.

Q>6: For a finer spin segmentation we still obtain the low-temperature stripe-like phase. A single first-order phase transition observed for $Q = 6$ is split into two transitions, at temperatures T_1 and T_2 with $T < T_1$ being the stripe-ordered phase. The intermediate phase $T_1 \leq T \leq T_2$ appears to be critical[4,10].

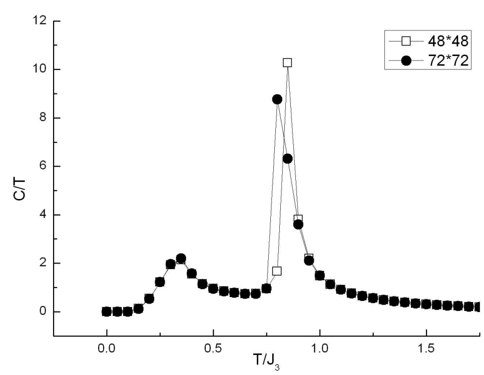

Fig. 2. Specific heat $C(T)/T$ for $Q = 12$, $H = H_3$, for 48×48 and 72×72 lattices. The two peaks are indicative of the presence of two phase transitions. The low temperature phase is given by the stripe configuration shown in Fig. 1.

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